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Weak localization of electromagnetic waves by densely packed many-particle groups: Exact 3D results

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Abstract

We use the superposition *T*-matrix method to solve the Maxwell equations and compute electromagnetic scattering characteristics of a 3D volume filled with densely packed, randomly distributed, wavelength-sized spherical particles. Our numerically exact data provide, for the first time, a direct demonstration of the onset and evolution of coherent backscattering with increasing number of particles and prove unequivocally that weak localization of electromagnetic waves can survive even in densely packed particulate media.

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Keywords: Coherent backscattering; Weak localization; Electromagnetic scattering

1. Introduction

The effect of weak localization (WL) (or coherent backscattering) of electromagnetic waves by discrete random media was predicted in [1] and has been the subject of active theoretical and laboratory research [2–7]. The origin of WL is illustrated in Fig. 1 which shows a random group of particles illuminated by a plane wave propagating in the direction $\hat{\mathbf{n}}_{\mathrm{ill}}$. If the observation direction $\hat{\mathbf{n}}_{\mathrm{obs}}$ is far from the exact backscattering direction given by $-\hat{\mathbf{n}}_{\mathrm{ill}}$, then the average effect of interference of conjugate scattered waves going through various strings of particles in opposite directions is zero, owing to randomness of particle positions. Consequently, the observer measures some average, incoherent intensity. However, at exactly the backscattering direction ($\hat{\mathbf{n}}_{\mathrm{obs}} = -\hat{\mathbf{n}}_{\mathrm{ill}}$), the phase difference between the conjugate paths involving any string of particles is identically equal to zero, and the interference is always constructive.

It is important to recognize that the very concepts of wave phase, reciprocity, and ladder and cyclical diagrams invoked to explain WL are implicitly based on the assumption that each particle in a particle string (Fig. 1) is located in the far-field zones of the previous and the following particle, so that a wave scattered by a particle develops into a transverse spherical wave by the time it reaches another particle [7]. This assumption is consistent with a number of successful laboratory measurements of WL for dilute particle suspensions and allows one to use the microphysical theory of WL valid in the limit of vanishing particle packing density and

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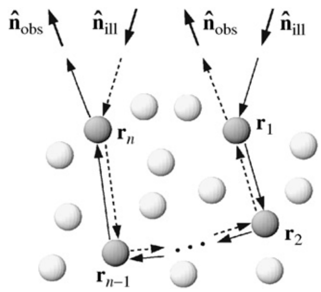

Fig. 1. Schematic explanation of WL. The direct (solid arrows) and reverse (dashed arrows) wave paths go through the same string of n particles, but in opposite directions.

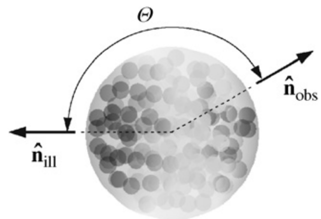

Fig. 2. Scattering geometry. In this case the large spherical domain is filled with 160 randomly positioned particles.

closely related to the microphysical theory of radiative transfer [7–10]. However, the criteria of far field scattering are rather stringent [7] and are often violated in the case of densely packed particles, thereby making more difficult and less definitive the interpretation of laboratory measurements of light backscattered by dense particle suspensions and particulate surfaces [11,12]. It is thus clear that quantitative analyses of backscattering measurements for densely packed particulate media must ultimately be based on direct solutions of the Maxwell equations.

To the best of our knowledge, an unequivocal demonstration of WL by 3D multi-particle groups based on a direct, numerically exact solution of the Maxwell equations has never been reported, which is explained by the extreme analytical and numerical complexity of such computations (e.g., [9,13,14]). However, the availability of the efficient and numerically exact superposition T-matrix approach [15,16] coupled with the rapidly increasing power of scientific workstations has made possible a direct demonstration of the onset and evolution of WL with increasing number of particles filling a finite 3D volume. This demonstration is the main objective of this paper.

2. Numerical results

As shown in Fig. 2, we assume that a number of identical small particles fill a spherical volume with a radius R much greater than the particle radius r. In our computations, we fixed the size parameter of the particles at $kr = 4$, where k is the wave number in the surrounding medium, whereas the size parameter of the spherical volume was fixed at $kR = 40$. The particle refractive index was set at 1.32, thus being representative of both water and water ice at visible wavelengths. The number of particles in the spherical volume, N, was varied between 1 and 160, thereby yielding particle volume concentrations ranging from 0.1% to 16%.

The randomness of particle positions inside the large spherical volume was simulated in two steps. First, we used a random-number generator to assign 3D coordinates to each of the N particles with a trial-and-error procedure ensuring that the particles do not overlap. Second, all group scattering characteristics were averaged over the uniform orientation distribution of the resulting N-particle configuration with respect to the laboratory coordinate system. This procedure yielded in effect an infinite number of random realizations of an N-particle group.

We assume that the large spherical volume is illuminated by a plane electromagnetic wave or a parallel quasi-monochromatic beam of light propagating in the direction $\hat{\mathbf{n}}_{\text{ill}}$. The angular distribution and polarization state of the light scattered by the entire N-particle group in random orientation is fully described by the so-called normalized Stokes scattering matrix. The latter specifies the transformation of the Stokes parameters I, Q, U, and V of the incident light into those of the light scattered in the observation direction $\hat{\mathbf{n}}_{\text{obs}}$ in the far-field zone of the entire spherical volume [16,17]:

$$\begin{bmatrix} I^{\text{obs}} \\ Q^{\text{obs}} \\ U^{\text{obs}} \\ V^{\text{obs}} \end{bmatrix} \propto \begin{bmatrix} a_1(\Theta) & b_1(\Theta) & 0 & 0 \\ b_1(\Theta) & a_2(\Theta) & 0 & 0 \\ 0 & 0 & a_3(\Theta) & b_2(\Theta) \\ 0 & 0 & -b_2(\Theta) & a_4(\Theta) \end{bmatrix} \begin{bmatrix} I^{\text{ill}} \\ Q^{\text{ill}} \\ U^{\text{ill}} \\ V^{\text{ill}} \end{bmatrix}. \tag{1}$$

In this equation, Θ is the scattering angle (Fig. 2), the Stokes parameters are defined with respect to the scattering plane, and the (1,1) element of the scattering matrix, called the phase function, is normalized:

$$\frac{1}{2}\int_0^{\pi} \mathrm{d}\Theta \, \sin\Theta a_1(\Theta) = 1. \tag{2}$$

The elements of the scattering matrix were computed using the highly efficient superposition T-matrix code described in [15,16]. The main advantage of this semi-analytical procedure is that the orientation-averaging step requires only a minor fraction of the total CPU time. The results of the extensive T-matrix computations are summarized in Fig. 3. Note that the specific block-diagonal structure of the scattering matrix in Eq. (1) follows from the T-matrix results and is largely caused by averaging over the uniform orientation distribution of the multi-particle groups and by sufficient randomness of particle positions within each group. All scattering matrix elements denoted by the zeros were found to be at least an order of magnitude smaller than the smallest of the non-zero elements (in the absolute-value sense).

3. Discussion

The phase function describes the angular distribution of the scattered intensity provided that the incident light is unpolarized. Fig. 3(a) clearly demonstrates several important effects of increasing the number of particles in the system. First, the constructive interference of light singly scattered by the component particles in the forward direction causes a strong forward-scattering enhancement [18] which eventually starts to resemble the diffraction peak typical of a solid particle with a size much larger than the wavelength [see the insert in Fig. 3(a)]. Second, the phase functions at scattering angles $30^\circ \leqslant \Theta \leqslant 170^\circ$ become progressively smooth and featureless, as a consequence of increasing amount of multiple scattering. Third, and most important from our perspective, the phase functions at scattering angles $\Theta > 170^\circ$ start to develop a backscattering enhancement which becomes quite pronounced for $N = 160$ [see Fig. 3(e)]. This feature has an angular width and an amplitude indicative of an interference origin [2–5] and, as we will further substantiate, is the most obvious manifestation of coherent backscattering.

The ratio a_2/a_1 is identically equal to one for scattering by a single sphere. Therefore, the rapidly growing deviation of this ratio from the value one for the multi-particle groups with $N \geqslant 5$ is a very sensitive indicator of the increasing effect of multiple scattering [Fig. 3(b)]. Similarly, $a_4(180^\circ)/a_1(180^\circ) = -1$ for single scattering by an isolated sphere, but multiple scattering in particle groups with $N \geqslant 5$ causes this quantity to grow quite significantly [Fig. 3(c)]. The degree of linear polarization of the scattered light for unpolarized incident light is given by the ratio $-b_1/a_1$. Panel 3(d) shows that the most obvious effect of increasing N is to smooth out the oscillations in the polarization curve for the single wavelength-sized sphere and, on average, to make polarization more neutral.

If the incident light is polarized linearly in the scattering plane then $Q^{\text{ill}} = I^{\text{ill}}$ and $U^{\text{ill}} = V^{\text{ill}} = 0$. The corresponding angular distributions of the co-polarized [i.e., $(I^{\text{obs}} + Q^{\text{obs}})/2$] and cross-polarized [i.e., $(I^{\text{obs}} - Q^{\text{obs}})/2$] scattered intensities are shown in Fig. 3(f) and (g), respectively. Similarly, Figs. 3(h) and (i) depict the "same-helicity" [i.e., $(I^{\text{obs}} + V^{\text{obs}})/2$] and "opposite-helicity" [i.e., $(I^{\text{obs}} - V^{\text{obs}})/2$] scattered intensities for the case of incident light polarized circularly in the counterclockwise direction when looking in

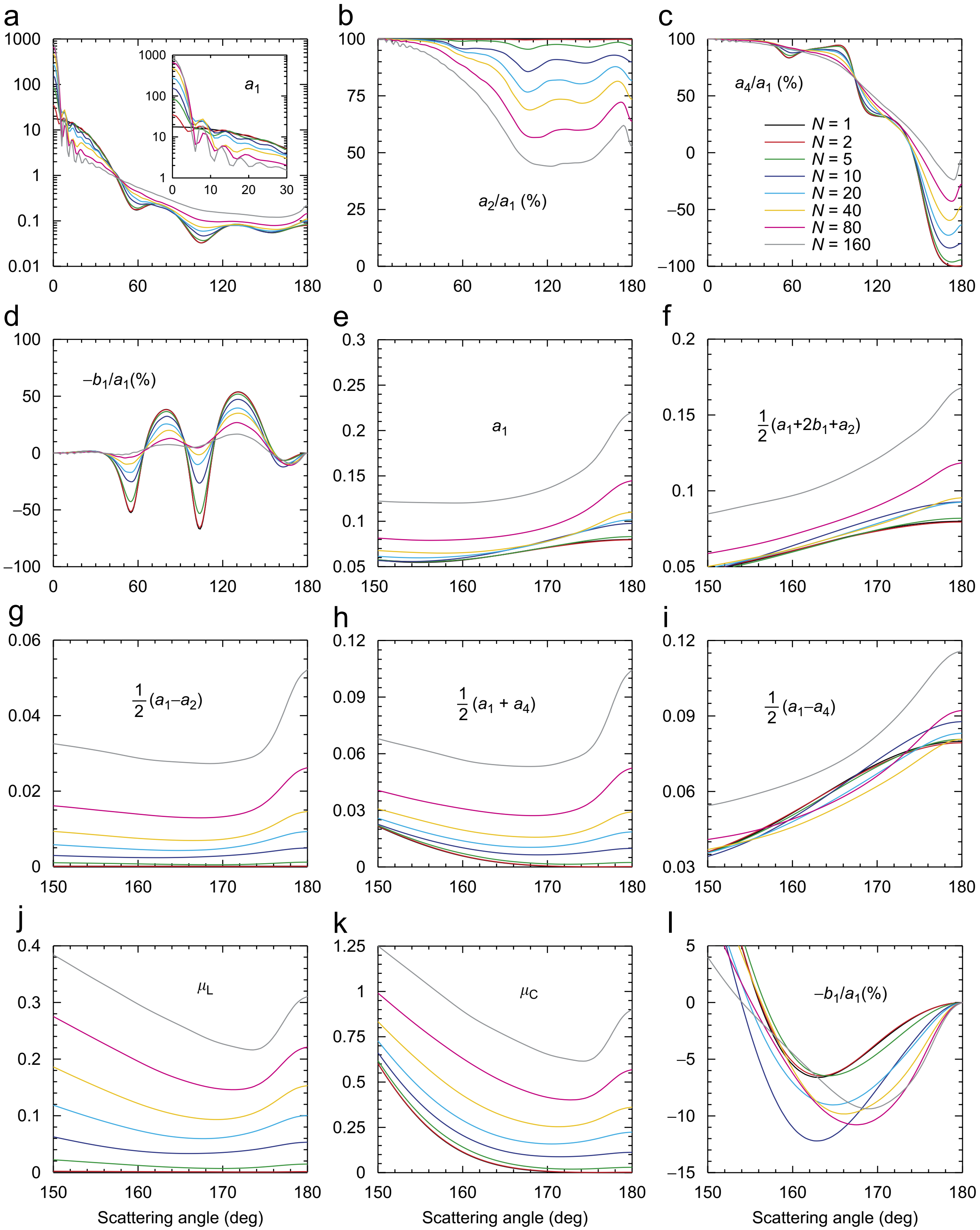

Fig. 3. Results of T-matrix computations.

the direction of propagation ($Q^{\rm ill} = U^{\rm ill} = 0$ and $V^{\rm ill} = I^{\rm ill}$). All of these quantities exhibit WL in the form of backscattering peaks rapidly growing in amplitude with N. The angular widths of these peaks are approximately the same, which testifies again to their common interference origin. The onset of coherent backscattering is especially demonstrative in Figs. 3(g) and (h) since the corresponding single-particle curves show no backscattering enhancement at all.

Figs. 3(j) and (k) depict the angular profiles of the so-called linear, $\mu_{\rm L}$, and circular, $\mu_{\rm C}$, polarization ratios defined as the ratio of the cross-polarized to co-polarized scattered intensities and the ratio of the same-helicity to the opposite-helicity scattered intensities, respectively. These quantities are widely used in radar and lidar remote sensing [7,19] because they vanish at the exact backscattering direction if multiple scattering is insignificant and the scattering particles are spherically symmetric. Our results show that multiple scattering causes an increasing deviation of $\mu_{\rm L}(180^\circ)$ and $\mu_{\rm C}(180^\circ)$ from zero, while WL causes pronounced backscattering peaks in the $\mu_{\rm L}$ and $\mu_{\rm C}$ angular curves.

All manifestations of multiple scattering and WL discussed so far are quite consistent, both qualitatively and semi-quantitatively, with those predicted by the asymptotic low-density microphysical theory [7–10]. This result indicates that even in densely packed particulate media, the waves scattered along strings of widely separated particles still provide a significant contribution to the total scattered signal and make quite pronounced the classical multiple-scattering and WL effects.

One manifestation of WL that is not seen in our results is the so-called polarization opposition effect (POE) [20]. However, POE causes a much more subtle polarization feature than the backscattering enhancement of intensity [10,20], and the former may be masked by the strong negative polarization branch at backscattering angles exhibited even by the single-particle curve [Fig. 3(l)]. Furthermore, it is still uncertain whether POE can be caused by wavelength-sized particles like the ones used in this study [21].

4. Conclusion

In summary, our exact T-matrix data provide a vivid demonstration of the onset and evolution of coherent backscattering with increasing number of particles and prove unequivocally that WL does occur even in densely packed particle groups. These results should have significant implications for particle characterization and remote sensing research [19,22–24]. Furthermore, the success of this study suggests that the T-matrix approach could eventually be used to model the onset of strong localization, which remains to be a rather ‘‘evasive’’ effect [25–29]. The advantage of the approach based on a direct and numerically exact solution of the Maxwell equations is that it can potentially yield all quantitative characteristics of a complex scattering system some (or most) of which may not be easy to observe and accurately measure. Another useful extension of our study would be to consider multiple scattering by nonspherical particles [30,31].

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